IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Appl. No.	:	10/571,508	**Confirmation No.:** 7832
Applicant (s)	:	Liping Zhang et al.	
Filed	:	March 10, 2006	
TC/A.U.	:	1625	
Examiner	:	Bernard I Dentz	
Title	:	A PROCESS FOR THE PRODUCTION OF ALKYLENE OXIDE USING A GAS-PHASE PROMOTER SYSTEM	
Docket No.	:	63261A	
Customer No.	:	00109	

Declaration Under 37 C.F. R. § 1.132

The undersigned, Dr. Liping Zhang, citizen of the United States of America and residing in the State of Texas, United States of America declare and say that:

1. I am a 1982 graduate of Tsing-hua University, Beijing, China with a Bachelor of Science degree in Chemical Engineering, a 1985 graduate of Tsing-hua University, Beijing, China with a Masters of Science degree in Chemical Engineering and a 1996 graduate of The Ohio State University, Columbus, Ohio, USA with a Doctor of Philosophy Degree in Chemical Engineering.

2. From April 1985 until April 1991, I was employed as an R&D Engineer at the Research Institute of Petroleum Processing, SINOPEC Co. in China. From September 1987 until December 1988 and then from May 1991 until September 1992, I was a Visiting Research Associate at The Ohio State University. From September 1992 through November 1996, I was a Graduate Research Associate with The Ohio State University.

3. In December 1996, I joined Union Carbide Corporation ("UCC") in South Charleston, West Virginia as a Senior Engineer. I held this position until March of 2000. At that time, I became a Project Scientist with UCC, and held this position until April, 2001, when UCC was acquired by The Dow Chemical Company ("TDCC"). After UCC became a wholly owned subsidiary of TDCC, I was named a Research Specialist. In March 2003, I was promoted to Senior Research Specialist; this is my role today. I

worked at the South Charleston site until 2008 when I relocated to TDCC's Freeport, Texas, location.

4. In my roles at UCC and TDCC, my responsibilities included leading the design, construction, and commissioning of the Ethylene Oxide ("EO") R&D ROTOBERTY® laboratory, the supervision of operation of ROTOBERTYs, autoclaves, pilot plant, and microreactor systems, conducting studies of catalyst formulation, catalyst preparation, catalyst characterization, and kinetics to improve the performance and fundamental understanding of EO catalysts. I also developed methods and experiments for EO catalyst characterization and kinetic studies, provided technical support for EO plants, and developed gas phase promoter strategies for EO plants.

5. I am currently an inventor on one US published application in the field of ethylene oxide production.

6. I have read the above referenced patent application, the Office Action dated February 13, 2009, and the prior art references, Thorsteinson et al., EP 425020 B1 ("Thorsteinson I"), Thorsteinson et al., EP 480537 ("Thorsteinson II"); and Hayden et al., US 5,387,751 cited by the Examiner.

7. Thorsteinson I does not disclose any catalysts which have produced 1.1 kilo metric tons of ethylene oxide per cubic meter of catalyst. In addition, I have determined the N*/Z* ratios for the relevant examples of Thorsteinson I, under the variety of conditions set forth in Thorsteinson I. The N*/Z* ratios for Thorsteinson I range from 0.18 to 2.99. Only three of 34 experiments use an N*/Z* ratio of between 0.4 to 1.0. Among these three experiments, i.e., Examples 28, 32, and 33, Example 33 had both N* (38) and Z* (80) values above the respective 20 and 40 values, as set forth in independent claim 1. Both Examples 28 and 33 are conducted under CO_2 concentrations of 3.0 mole %. There is no experiment in which all variables were constant except for N*/Z*. Thorsteinson I contains no teaching of the benefit of the claimed N*/Z* ratio for fresh catalysts , let alone for catalysts which have produced the claimed amount of ethylene oxide. See Table 1, attached to this declaration, for a list of the experimental conditions.

<u>Examples 1-6 and 7-12</u>

N*/Z* Condition I: 0.26

N^*/Z^* Condition II: 0.39

<u>Examples 13-15</u>

N^*/Z^*: 0.18

<u>Examples 16-21</u>

N^*/Z^* O_2 Condition: 0.26

N^*/Z^* Air Condition: 0.22

<u>Examples 22-27</u>

N^*/Z^* 0.26

<u>Examples 28-31</u>

N^*/Z^* Condition I: 0.93

N^*/Z^* Condition II: 0.27

N^*/Z^* Condition III: 2.99

N^*/Z^* Condition IV: 1.36

<u>Examples 32-34</u>

N^*/Z^* Air Condition: 0.51, where inlet C2H4% = 12

N^*/Z^* O_2 Condition I: 0.47, where N^* =37.7 and Z^* = 80.0

N^*/Z^* O_2 Condition II: 0.38

8. Similarly, Thorsteinson II does not disclose any catalysts which have produced 1.1 kilo metric tons of ethylene oxide per cubic meter of catalyst. In addition, I have determined the N^*/Z^* ratios for the relevant examples of Thorsteinson II. The N^*/Z^* ratios range from .20 to .48. Only 5 of 13 experiments in Thorsteinson II use an N^*/Z^* ratio between 0.4 and 1.0. For Catalyst Nos. 7, 8, and 9, the Z^* value is 66.7, far above the value of 40 set forth in independent claim 1. There is no experiment in which all variables were constant except for N^*/Z^*. Thorsteinson II contains no teaching of the benefit of the claimed N^*/Z^* ratio for fresh catalysts, let alone for catalysts which have

produced the claimed amount of ethylene oxide. Instead, analysis of the data in Table 2 of Thorsteinson II shows that for younger catalysts, a lower N*/Z* ratio provides better performance. Compare Catalyst 4 at 20 days, having an N*/Z* ratio of 0.35, with an efficiency of 85.8%, % EO of 1.98, and temperature of 255C with Catalyst Nos. 1-3, which have an N*/Z* ratio of greater than 0.4. The increase in the N*/Z* ratio is coupled with a decrease in efficiency, or decrease in %EO as compared to Catalyst No. 4.

Catalyst No.	N*	Z*	N*/Z*
1	6.9	14.7	0.47
1	7.2	15.0	0.48
1	7.1	16.3	0.44
2	7.2	15.7	0.46
3	9.2	20.7	0.45
4	6.9	20.0	0.35
5	4.3	13.3	0.33
6	13.0	66.7	0.20
7	13.0	66.7	0.20
8	13.0	66.7	0.20
9	13.0	66.7	0.20
10, Condition I	4.3	16.7	0.26
10, Condition II	10.4	33.3	0.31

9. Similarly, Hayden does not disclose any catalysts which have produced 1.1 kilo metric tons of ethylene oxide per cubic meter of catalyst. In addition, I have determined the N*/Z* ratios for the relevant examples, under the varying conditions of Hayden. Where greater than zero, the N*/Z* ratios for Hayden range from 0.0006 to 0.099. Thus, none are within the claimed range. In Examples 13, 20, 29, 34 and 15, Z* is in excess of 66.7, far above the value of 40 set forth in independent claim 1. Hayden

contains no teaching of the benefit of the claimed N*/Z* ratio for fresh catalysts, let alone for catalysts which have produced the claimed amount of ethylene oxide. See Table 2, attached to this declaration, for the experimental conditions.

10. The following experiment demonstrates that a catalyst which has generated more than 1.1 kilo metric tons of ethylene oxide per m3 of catalyst benefits from operating within the claimed N*/Z* ratio.

Tubular fixed bed reactors contain a mixture of catalysts which have been used to produce ethylene oxide. The N*/Z* ratio was decreased from greater than 1 to 0.6. Catalyst efficiency (selectivity) improved by more than 1% without the loss of catalyst activity.

Properties of fresh catalysts in the reactor:

	Average	High	Low
Silver content (wt% by titration)	>30	>30	>30
Promoter	Potassium	Potassium	Potassium

Reactor inlet conditions:

Pressure (kPa, absolute)	2270 to 2310
Gas hourly space velocity (h-1)	6100 to 6900
Ethylene concentration (mol %)	21 to 27.5
Ethane concentration (mol %)	0 to 0.03
Oxygen concentration (mol %)	3 to 8.7
Carbon dioxide concentration (mol %)	0.2 to 0.5
Water concentration (mol %)	0.8 to 1.2
Ethylene dichloride (ppm)	0.3 to 1.1
Vinyl chloride (ppm)	0.5 to 2.0
Ethyl chloride (ppm)	0.0 to 0.03
Ammonia feed (ppm)	3.7 to 12.8
Total Nox (ppm) (estimated value based on material balance)	0.4 to 1.3
Balance gas: methane + nitrogen + argon	

The work rate (also referred to as the catalyst ethylene oxide loading) was between 150 to 270 (kgEO/h/m3 catalyst). The reactor coolant temperature ranged from 218 to 240°C.

The attached plots contain the "normalized" relative efficiency and "normalized" relative coolant temperature vs. cumulative EO loading. The normalized efficiency values were obtained by using the catalyst kinetic correlations to normalize the raw efficiency values to a reference work-rate of 238 kgEO/h/m3. The normalized efficiency values were then converted to the relative efficiency values by subtracting a constant reference efficiency number to convenient the comparison. The normalized coolant temperature values were obtained by using the catalyst kinetic correlations to normalize the raw coolant temperature values to a reference 238 kgEO/h/m3 work-rate and a reference 8.5% (mole) inlet oxygen concentration. The normalized coolant temperature values were then converted to the relative coolant temperature values by subtracting the same reference coolant temperature number to convenient the comparison.

Fig. 1 depicts N*/Z* on the left y-axis, N* or Z* on the right y-axis, and cumulative catalyst loading (kt EO/m3) along the x-axis.

Fig. 2 depicts N*/Z* on the left y-axis, relative efficiency (%) on the right y-axis and cumulative catalyst loading (kt EO/m3) along the x-axis.

Fig. 3 depicts Fig. 2 depicts N*/Z* on the left y-axis, normalized coolant temperature (C) on the right y-axis and cumulative catalyst loading (kt EO/m3) along the x-axis.

The figures demonstrate that operating the process at an N*/Z* ratio of between 0.4 to 1.0 for a catalyst which has produced in excess of 1.1 kilo metric tons of ethylene oxide per m3 of catalyst provides a selectivity increase but does not affect activity. Further, the figures demonstrate that the benefit of operating at the claimed N*/Z* ratio range is greater for the catalyst which has produced in excess of 1.1 kilo metric tons of ethylene oxide per m3 of catalyst than the benefit to a catalyst which has produced less than that volume of ethylene oxide.

Data between 0.42 to 0.51 ktEO/m3 cumulative catalyst loading were not plotted due to some uncertainty on measurements accuracy.

Fig. 1

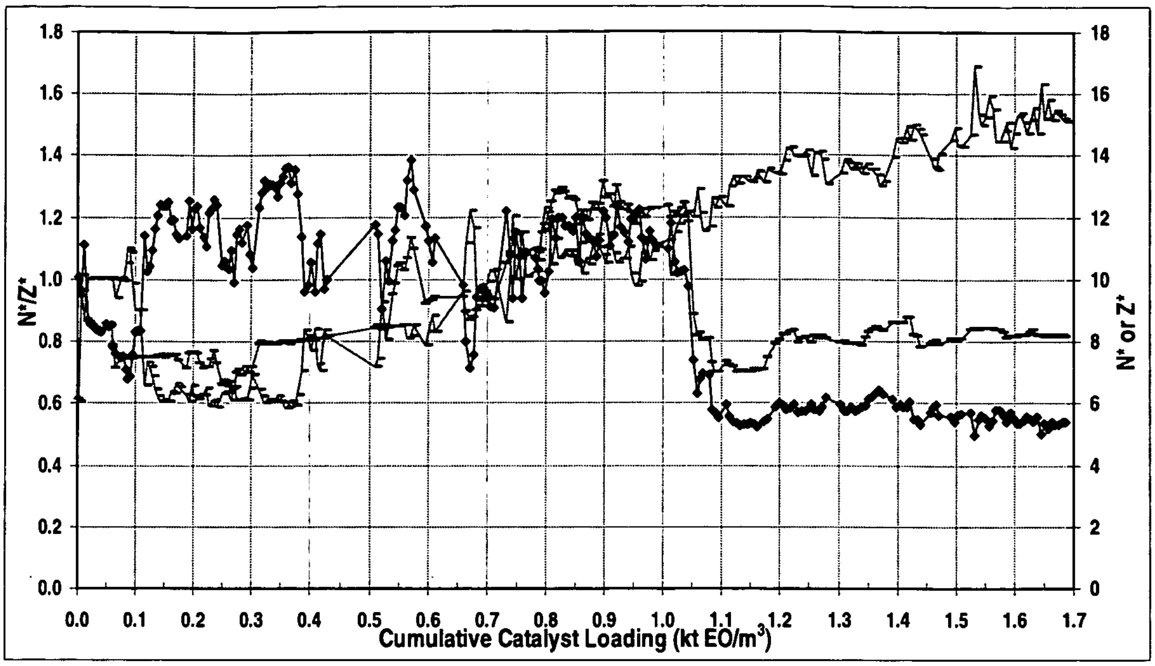

1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
N*/Z*
18
16
14
12
10
8
6
4
2
0
N* or Z*
0.0 0.1 0.2 0.3 0.4 0.5 0.6 0.7 0.8 0.9 1.0 1.1 1.2 1.3 1.4 1.5 1.6 1.7
Cumulative Catalyst Loading (kt EO/m³)

Fig. 2

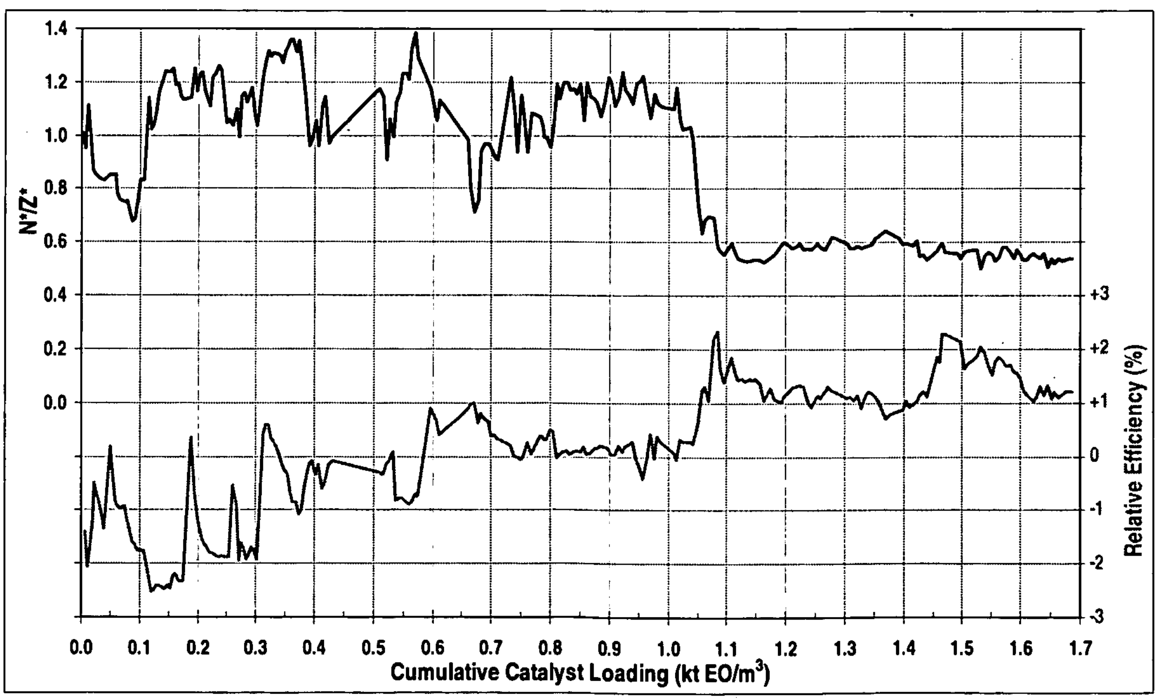
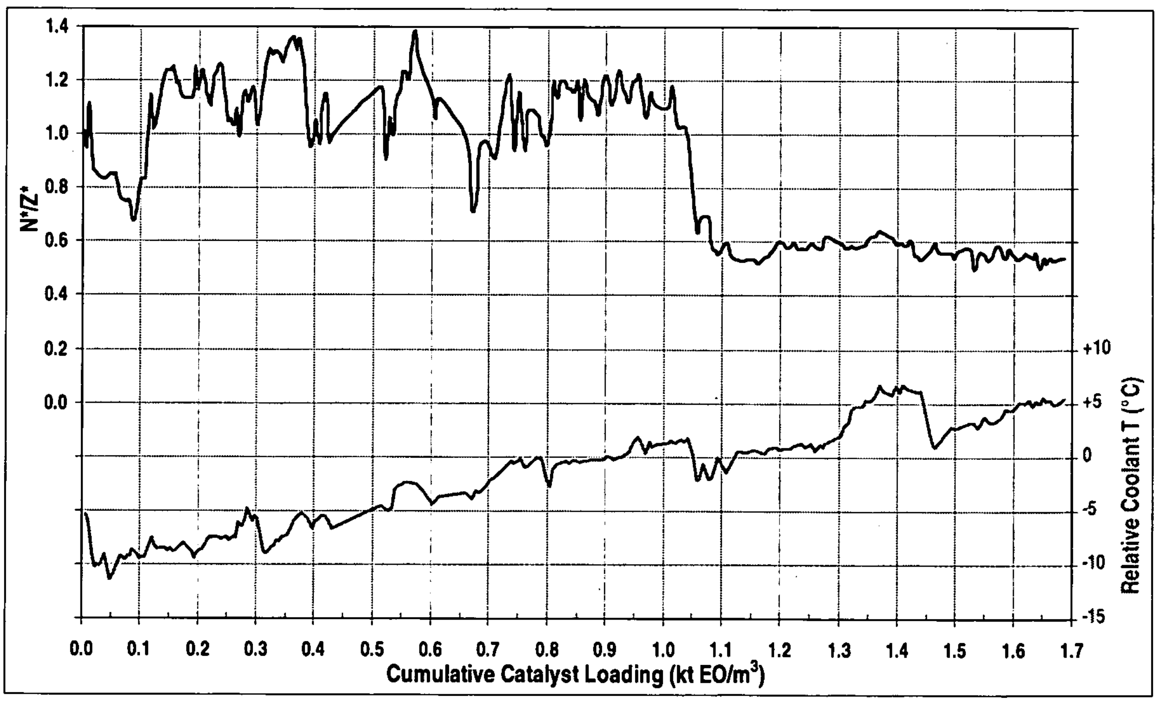

1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
N*/Z*
+3
+2
+1
0
-1
-2
-3
Relative Efficiency (%)
0.0 0.1 0.2 0.3 0.4 0.5 0.6 0.7 0.8 0.9 1.0 1.1 1.2 1.3 1.4 1.5 1.6 1.7
Cumulative Catalyst Loading (kt EO/m³)

Fig. 3



11. I declare further that all statements made herein of my knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title

18 of the United States Code, and that willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Respectfully submitted,

Liping Zhang

Table 1

Examples 1-6 and 7-12

		P		NO	N*	ECL	C2H4	C2H6	Z*	N*/Z*	CO2	O2	EO out	Day	T	GHSV
		psig	kPa	ppm		ppm	%	%			%	%	%		°C	h-1
Example 1	Condition I	275	1997	5	4.3	5	30.0	0.0	16.7	0.26	0	8.0	0.90	20	240	8000
	Condition II	275	1997	15	13.0	20	30.0	0.3	33.3	0.39	3.0	8.0	0.17	20	270	8000
Example 2	Condition I	275	1997	5	4.3	5	30.0	0.0	16.7	0.26	0	8.0	1.60	26	240	8000
	Condition II	275	1997	15	13.0	20	30.0	0.3	33.3	0.39	3.0	8.0	0.16	26	270	8000
Example 3	Condition I	275	1997	5	4.3	5	30.0	0.0	16.7	0.26	0	8.0	1.60	23	240	8000
	Condition II	275	1997	15	13.0	20	30.0	0.3	33.3	0.39	3.0	8.0	0.40	23	270	8000
Example 4	Condition I	275	1997	5	4.3	5	30.0	0.0	16.7	0.26	0	8.0	1.60	35	240	8000
	Condition II	275	1997	15	13.0	20	30.0	0.3	33.3	0.39	3.0	8.0	0.67	35	270	8000
Example 5	Condition I	275	1997	5	4.3	5	30.0	0.0	16.7	0.26	0	8.0	1.50	48	240	8000
	Condition II	275	1997	15	13.0	20	30.0	0.3	33.3	0.39	3.0	8.0	0.83	48	270	8000
Example 6	Condition I	275	1997	5	4.3	5	30.0	0.0	16.7	0.26	0	8.0	1.60	40	240	8000
	Condition II	275	1997	15	13.0	20	30.0	0.3	33.3	0.39	3.0	8.0	0.80	40	270	8000
Example 7	Condition I	275	1997	5	4.3	5	30.0	0.0	16.7	0.26	0	8.0	1.69	2	240	8000
	Condition II	275	1997	15	13.0	20	30.0	0.3	33.3	0.39	3.0	8.0	1.71	9	270	8000
Example 8	Condition I	275	1997	5	4.3	5	30.0	0.0	16.7	0.26	0	8.0	1.82	2	240	8000
	Condition II	275	1997	15	13.0	20	30.0	0.3	33.3	0.39	3.0	8.0	1.97	9	270	8000
Example 9	Condition I	275	1997	5	4.3	5	30.0	0.0	16.7	0.26	0	8.0	1.85	1	240	8000
	Condition II	275	1997	15	13.0	20	30.0	0.3	33.3	0.39	3.0	8.0	2.00	12	270	8000
Example 10	Condition I	275	1997	5	4.3	5	30.0	0.0	16.7	0.26	0	8.0	2.03	1	240	8000
	Condition II	275	1997	15	13.0	20	30.0	0.3	33.3	0.39	3.0	8.0	2.07	4	270	8000
Example 11	Condition I	275	1997	5	4.3	5	30.0	0.0	16.7	0.26	0	8.0	1.54	10	240	8000
	Condition II	275	1997	15	13.0	20	30.0	0.3	33.3	0.39	3.0	8.0	1.61	13	270	8000
Example 12	Condition I	275	1997	5	4.3	5	30.0	0.0	16.7	0.26	0	8.0	2.12	7	240	8000
	Condition II	275	1997	15	13.0	20	30.0	0.3	33.3	0.39	3.0	8.0	1.96	9	270	8000

Examples 13-15

		P		NO	N*	ECL	C2H4	C2H6	Z*	N*/Z*	CO2	O2	EO out	Day	T	GHSV
		psig	kPa	ppm		ppm	%	%			%	%	%		°C	h-1
Example 13		275	1997	7	6.1	10	30.0	0.0	33.3	0.18	3.0	8.0	1.30		236	8000
Example 14		275	1997	7	6.1	10	30.0	0.0	33.3	0.18	3.0	8.0	1.45		236	8000
Example 15		275	1997	7	6.1	10	30.0	0.0	33.3	0.18	3.0	8.0	1.10		236	8000

Examples 16-21

		P		NO	N*	ECL	C2H4	C2H6	Z*	N*/Z*	CO2	O2	EO out		T	GHSV
		psig	kPa	ppm		ppm	%	%			%	%	%		°C	h-1
Example 16	O2 Condition	275	1997	5	4.3	5	30.0	0.0	16.7	0.26	0.0	8.0	1.10		240	8000
Example 18	O2 Condition	275	1997	5	4.3	5	30.0	0.0	16.7	0.26	0.0	8.0	1.90		240	8000
Example 17	Air Condition	275	1997	8	6.9	12	8.0	0.3	31.6	0.22	4.0	6.0	0.40		270	8000
Example 19	Air Condition	275	1997	8	6.9	12	8.0	0.3	31.6	0.22	4.0	6.0	0.90		270	8000
Example 20	Air Condition	275	1997	8	6.9	12	8.0	0.3	31.6	0.22	4.0	6.0	0.45		270	8000
Example 21	Air Condition	275	1997	8	6.9	12	8.0	0.3	31.6	0.22	4.0	6.0	0.90		270	8000

Examples 22-27

		P		NO	N*	ECL	C2H4	C2H6	Z*	N*/Z*	CO2	O2	EO out		T	GHSV
		psig	kPa	ppm		ppm	%	%			%	%	%		°C	h-1
Example 22		275	1997	5	4.3	5	30.0	0.0	16.7	0.26	0	8	1.04		223	8000
Example 23		275	1997	5	4.3	5	30.0	0.0	16.7	0.26	0	8	1.51		218	8000
Example 25		275	1997	5	4.3	5	30.0	0.0	16.7	0.26	0	8	1.30		220	8000
Example 24		275	1997	5	4.3	5	30.0	0.0	16.7	0.26	3	8	1.50		220	8000
Example 26		275	1997	5	4.3	5	30.0	0.0	16.7	0.26	3	8	1.12		220	8000
Example 27		275	1997	5	4.3	5	30.0	0.0	16.7	0.26	3	8	1.95		220	8000

Examples 28-31

		P		NO	N*	ECL	C2H4	C2H6	Z*	N*/Z*	CO2	O2	EO out	Eff	T	GHSV
		psig	kPa	ppm		ppm	%	%			%	%	%	%	°C	h-1
Example 28		275	1997	16	13.9	7.5	20.0	0.3	15.0	0.93	3.0	7.5	1.00	88	250	6200
Example 29		220	1618	20	14.1	23.0	24.0	0.2	52.3	0.27	1.0	8.0	0.35	88	260	4700
Example 30		300	2170	38	35.8	6.0	30.0	0.2	12.0	2.99	3.0	8.5	2.22	87	243	3600
Example 31		240	1756	52	39.7	14.0	28.0	0.2	29.2	1.36	5.0	8.5	2.30	86	258	3300

Examples 32-34

		P		NO	N*	ECL	C2H4	C2H6	Z*	N*/Z*	CO2	O2	EO out	Eff	T	GHSV
		psig	kPa	ppm		ppm	%	%			%	%	%	%	°C	h-1
Example 32	Air Condition	220	1618	15	10.6	15	12.0	0.6	20.8	0.51	2.0	8.0		82.0	265	5300
Example 33	O2 Condition	300	2170	40	37.7	40	30.0	0.2	80.0	0.47	3.0	8.5		88.0	245	3600
Example 34	O2 Condition	300	2170	6	5.7	9	30.0	0.3	15.0	0.38	0.5	8.5		89.0	220	6600

Table 2

Example No.	P (psia)	C2H4 (%)	C2H6%	VCL ppm	NO ppm	N*	Z*	N*/Z*
2	240	30	0	10	3	2.16	33.3	0.065
4	20	30	0	10	5	0.30	33.3	0.009
9	240	30	0	10	2	1.44	33.3	0.043
13	19.7	30	0	20	10	0.59	66.7	0.009

20	14.7	30	0	20	12	0.53	66.7	0.008
25	240	30	0	10	2	1.44	33.3	0.043
26	240	31	0.6	17	1.5	1.08	18.7	0.058
29	19.7	30	0	20	1	0.06	66.7	0.001
34	19.7	30	0	20	1	0.06	66.7	0.001

Example Nos. 10 and 11	P (psia)	C2H4 (%)	C2H6%	VCL ppm	NO ppm	N*	Z*	N*/Z*
Catalyst No. 9	240	30	0	10	2	1.44	33.3	0.043
Catalyst No. 10	240	30	0	6.5	1	0.72	21.7	0.033
Catalyst No. 11	240	30	0	7	1	0.72	23.3	0.031
Catalyst No. 12	240	30	0	7.5	0.5	0.36	25.0	0.014
Catalyst No. 13	240	30	0	8	2	1.44	26.7	0.054
Catalyst No. 14	240	30	0	9	1	0.72	30.0	0.024
Catalyst No. 15	240	30	0	10	1	0.72	33.3	0.022
Catalyst No. 16	240	30	0	7	0.5	0.36	23.3	0.015
Catalyst No. 17	240	30	0	9	1	0.72	30.0	0.024
Catalyst No. 18	240	30	0	**10**	**0.9**	0.65	33.3	0.019
Catalyst No. 19	240	30	0	10	0.9	0.65	33.3	0.019
Catalyst No. 20	240	30	0	2	0.7	0.50	6.7	0.075
Catalyst No. 21	240	30	0	2	0.7	0.50	6.7	0.075

Catalyst No. 22	240	30	0	2	0.7	0.50	6.7	0.075
Catalyst No. 23	240	30	0	2	0.7	0.50	6.7	0.075
Catalyst No. 24	240	30	0	4	1.5	1.08	13.3	0.081
Catalyst No. 25	240	30	0	10	1	0.72	33.3	0.022
Catalyst No. 26	240	30	0	6	1.4	1.01	20.0	0.050
Example No. 15	P (psia)	C2H4 (%)	**N-compounds**	VCL ppm	**N- ppm**	N*	Z*	N*/Z*
Catalyst No. 31	14.7	30	none	20	0	0.00	66.7	0.000
Catalyst No. 31	14.7	30	NO	20	20	0.88	66.7	0.013
Catalyst No. 31	14.7	30	NO2	20	20	0.88	66.7	0.013
Catalyst No. 31	14.7	30	NH3	20	150	6.61	66.7	0.099
Catalyst No. 31	14.7	30	C2H4(NH2)2	20	70	3.08	66.7	0.046
Catalyst No. 32	14.7	30	none	20	0	0.00	66.7	0.000
Catalyst No. 32	14.7	30	NO	20	10	0.44	66.7	0.007
Catalyst No. 32	14.7	30	NO2	20	10	0.44	66.7	0.007
Catalyst No. 32	14.7	30	NH3	20	150	6.61	66.7	0.099
Catalyst No. 32	14.7	30	C2H4(NH2)2	20	40	1.76	66.7	0.026
Catalyst No. 32	14.7	30	CH3CN	20	20	0.88	66.7	0.013
Example No. 18	P (psia)	C2H4 (%)	C2H6%	VCL ppm	NO ppm	N*	Z*	N*/Z*

Catalyst No. 35 (at 7 ppm vinyl chloride)	240	30	0	7	1.5	1.08	23.3	0.046
Catalyst No. 35 (at 6 ppm vinyl chloride)	240	30	0	6	1.5	1.08	20.0	0.054
Catalyst No. 35 (at 5 ppm vinyl chloride)	240	30	0	5	1.5	1.08	16.7	0.065
Example No. 22	P (psia)	C2H4 (%)	C2H6%	VCL equivalent ppm	NO ppm	N*	Z*	N*/Z*
Catalyst No. 38 (10 ppm vinyl chloride)	19.7	30	0	10	10	0.59	33.3	0.018
Catalyst No. 38 (10 ppm dichloroethane)	19.7	30	0	10	10	0.59	33.3	0.018
Catalyst No. 38 (50 ppm methyl chloride)	19.7	30	0	10	10	0.59	33.3	0.018
Catalyst No. 38 (4 ppm 2-chloropropane)	19.7	30	0	10	10	0.59	33.3	0.018